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Sixth Quarterly Technical Report

Analysis and Evaluation of Technical Data
on the
Photochromic and Non-Linear Optical
Properties of Materials

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BACKGROUND

The primary purpose of this relatively small contractual effort is to provide technical support to DARPA and CNVEO at Fort Belvoir in evaluating and assessing data on materials, especially polymers, that may be useful in the development of optical limiters and switches. The Principal Investigator was appointed by the former DARPA program manager for this project to be part of an ad hoc Technical Advisory Committee. Due to changes in the overall direction of the program, this committee has not

formally met during this quarter. *Keywords: optics, Materials, Ablative mirror-fuse system, Optical device, Ablative polymer layer, Photochromic and non-linear Optical*

RECENT PROGRESS

Attached to the third quarterly report, September 1, 1989, was a Working Draft Paper evaluating the feasibility of developing an ablative mirror-fuse system to be placed at the focal plane of an optical device. Incoming light first passes through a focus at the mirror optical-fuse before being recollimated prior to entering the observers eye. Focused light, if of sufficient intensity, would ablate the reflective layer on the mirror, disrupting the optical path. This draft paper proposed the use of a thermally unstable material (thermochemical enhancer), probably a thermally unstable polymer or an energetic material doped in a polymer, placed under a thin (100-300 nm) reflective layer. This thin ablative polymer layer would serve to the lower threshold for mirror-fuse activation. Further analysis of this concept is underway. Emphasis is being given to theoretically optimizing the reflective material as well as the thermochemical enhancer layer. Attached to this, the sixth quarterly report, is a Working Draft containing further data required for the development of a suitable optical mirror fuse system.



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The Principal Investigator attended the 199th National Meeting of the American Chemical Society and participated in the "Symposium on New Materials for Nonlinear Optics." The Principal Investigator was also co-author of a paper on "Third Order Optical Nonlinearities of Bis-Phthalocyanines." This research effort was not funded as part of this contract. Several papers presented at this meeting are worthy of special note and further investigation. These may be summarized as follows:

1. The majority of papers presented involved second order effects rather than third order.
2. Most of the NLO materials reported at the symposium were being developed for use in optical communications and computing. Requirements for this application are much less stringent than those required for eye protection devices. Figures of merit are much higher for development of an effective eye protection system.
3. Several third order materials were reported but most papers focused on synthesis and theory rather than experimental measurement of nonlinearity.
4. The largest $\chi^{(3)}$'s reported were in the range of 10^{-10} - 10^{-12} esu. Some were clearly resonance measurements. The largest non-resonant $\chi^{(3)}$'s reported were for transition metal and rare earth phthalocyanines currently being developed at the Naval Research Laboratory.
5. Interesting work was reported on polysilanes, germanes, boron-based molecules and phosphorous-nitrogen compounds.

6. R. D. Miller of IBM reported a χ^3 of $10^{-11} - 10^{-12}$ esu for a polysilane/germane polymer. This system is somewhat analogous to classical semiconductors.
7. Of special note is work currently in progress by K. R. Seddon (University of Sussex, UK) involving a transparent polymer host in which NLO (organic) micro-crystal guests were grown in place using a modified zone-melting technique. Their work has concentrated on second-order effects, however the technique could prove useful as well for third order materials where alignment and order could enhance non-linear behavior.

The second quarterly report May 31, 1989 had appended to it an exhaustive survey of the literature listing materials (especially organic) and reported values for their χ^2 and χ^3 . Much of the data that appeared in this appendix has since been published as a formal technical report through the Naval Research Laboratory where the Principal Investigator is also jointly employed as an Intermittent Senior Research Chemist GM-15. This report appears as NRL Memorandum Report 6482 by M. E. Boyle and R. F. Cozzens dated June 12, 1989. Copies are available through the Naval Research Laboratory, Washington, D. C.

The Principal Investigator plans to continue assessing new third order material as data is reported as well as further work on the theory and application of possible mirror optical-fuse systems.

**APPENDIX
TO
SIXTH QUARTERLY REPORT**

WORKING DRAFT

**FURTHER DATA AND ASSESSMENTS
OF
REFLECTIVE LAYERS
FOR A
MIRROR OPTICAL FUSE**

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To serve as a practical eye protection element, a device must be capable of responding at incident energy densities lower than the damage threshold of the eye. Commonly accepted eye damage thresholds indicate that the device must be able to respond to about 0.1 uJ/cm^2 of incident light in a nonosecond time frame (approximately 100 W cm^2 on a power scale.)^{1,2} To overcome this sensitivity issue, most proposed passive eye protection devices require a focal plane where the incident light is concentrated onto an active surface. Two examples of such an approach are the "optical switch" and the "optical fuse". In the "switch" system, a nonlinear optical material whose index of refraction or absorption/reflectivity is affected by the focussed incident light is employed. Many researchers are working on developing nonlinear optical materials for use in such a device. However, while dramatic improvements in material response are being reported, the nonlinear optical response of state-of-the-art materials is still inadequate for eye protection and/or is only active over a limited frequency range. The "mirror-fuse" system employs a thin sacrificial reflector at the focal plane, designed to fail (i.e. ablate) at an irradiance threshold below that for eye damage.

A schematic view of a generic sacrificial mirror device is depicted in Figure 1. The incident radiation passes through an objective lens which focusses the light onto the sacrificial mirror. At low incident intensities, the sacrificial mirror reflects the incident light towards the eye by way of a correcting lens system. At, sufficiently high incident intensities, the sacrificial mirror ablates, interrupting the optical path to the eye. The incident energy is then absorbed by the beam dump which could simply be the mirror substrate or an absorptive block behind a transparent mirror substrate. The beam dump can also be envisioned as being a detector which, once triggered,

activates an additional, independent protection system. Care must be taken to insure that the beam dump (mirror substrate) is itself not sufficiently reflective, while still optically flat, that it directs damaging light intensity to the eye. One way to avoid this situation is to apply the reflective optical fuse layer (mirror) to the back surface of a transparent support and place an absorbing beam dump behind and at an angle to that of the reflective coating. In this configuration, the destruction of the mirror results in a direct path to the beam dump with a minimum amount of reflected light directed back towards the eye.

Since the region of the mirror that is damaged by the incident radiation is small, a few microns in area, the mirror needs to be moved only a small distance in order to restore vision. With an appropriate design, Figure 2, it should be possible to rapidly mechanically reposition a new location on the sacrificial mirror into the optic path. That is, to reset the "optical circuit breaker" and thereby quickly reestablish vision.

To protect the eye without the use of a focal plane requires a material that can respond to an incident energy density less than 0.1 uJ cm^2 in a nanosecond time frame. Most metal mirror materials have intrinsic damage thresholds, for an optically pure surface, in the range of 0.5 to 4 J/cm^2 for nsec pulses (Table 1).³ Thus, the focal plane is seen to be an indispensable component of the sacrificial mirror concept. The use of a mirror design requires an optical system anyway, so the inclusion of an focal plane is not an unreasonable restraint.

The maximum damage threshold of the sacrificial mirror estimated in the preceding section for successful eye protection, 0.1 J/cm^2 , is several times more sensitive than the measured damage threshold of pure, reflective, (i.e ca. 95% reflective) aluminum, one of the most easily ablated of several common mirror materials (see Table 1.) In order to assess the potential for achieving the maximum damage threshold in common as well as uncommon elemental metal mirror materials it is first necessary to identify the important material parameters and how they affect the damage threshold.

Laser-produced damage in thin films is generally due to either dielectric breakdown induced by the electric field of the laser radiation or by the thermal absorption of laser energy by the film. The ablative mirror materials under consideration here are metals and therefore exclude the more wavelength-selective dielectric mirrors.

Laser-produced damage in metallic thin film mirrors such as Al, Ag, Cr, Cu, Au, Mo and Rh, should occur predominately by a thermal absorption mechanism. Thermal absorption may be increased over that of the optically pure reflective metal by various means of artificial "contamination" of the reflector, such as co-vapor deposition of other metals or carbon to increase absorptivity and, of course, decreased reflectivity. A suitable eye protection device for use in the field would still probably be acceptable with 50% or less reflectivity. Typical sun glasses often absorb over 90% of the visible ambient light. By increasing absorptivity, the threshold for ablation shown

in Table I is proportionately lowered. Aluminum with its absorptivity artificially increased to some acceptable level (ca. 50%) by controlled contamination of the thin reflective layer appears to be one of the most suitable reflectors for use in an optical mirror-fuse system.

The threshold for fuse failure may be lowered further by application of a thermally unstable energetic layer under the thin metal reflector. At some energy threshold, the substrate layer could be made to rapidly decompose and destroy the reflective (mirror) layer. Investigation of possible materials and compositions for this thermodynamically enhanced ablative layer are currently in progress.

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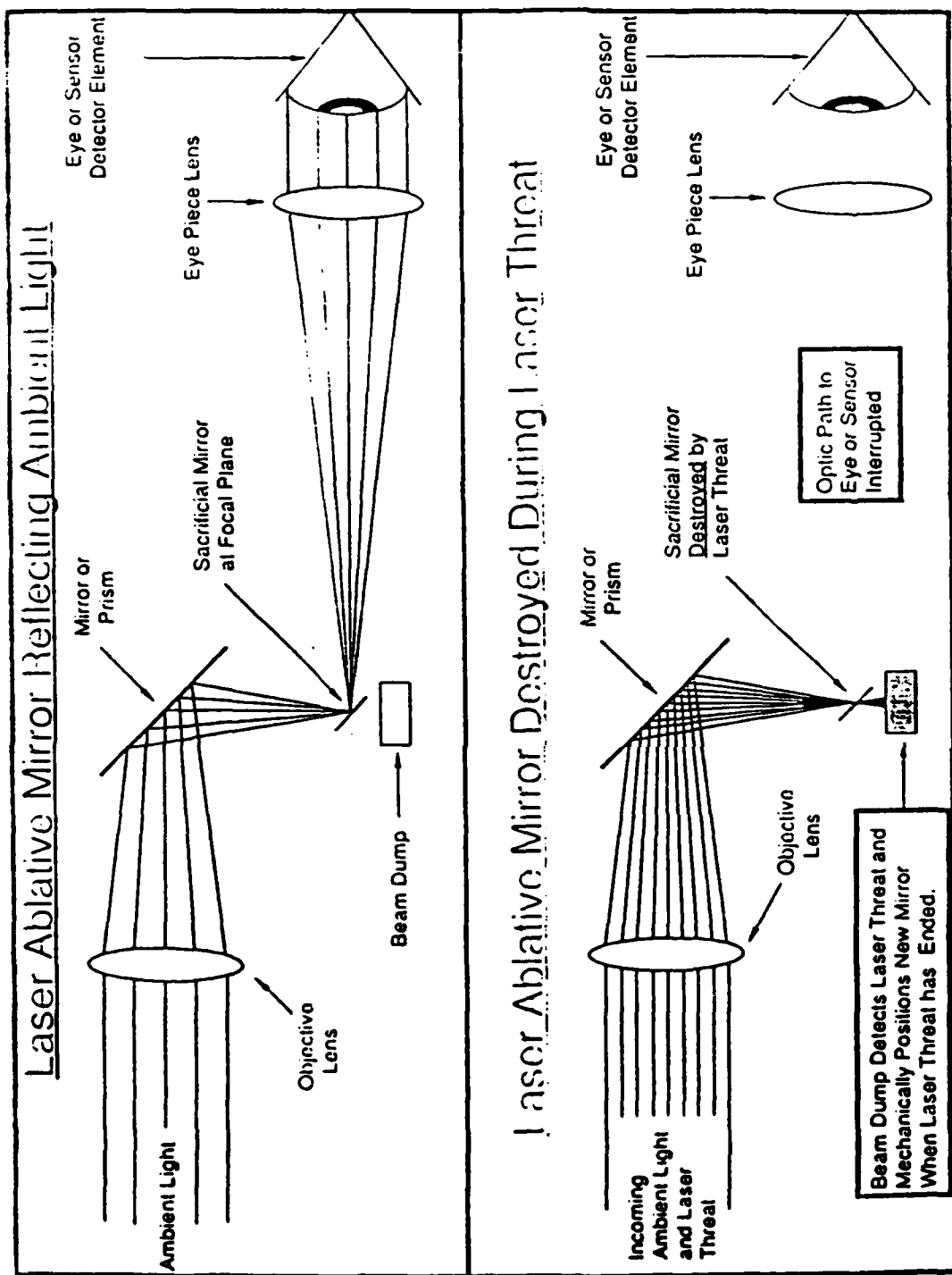


Figure 1a. A schematic diagram of the basic optical system for a mirror/optical-fuse device for eye protection against laser irradiation.

Laser Ablative Mirror: Parameter Schematic

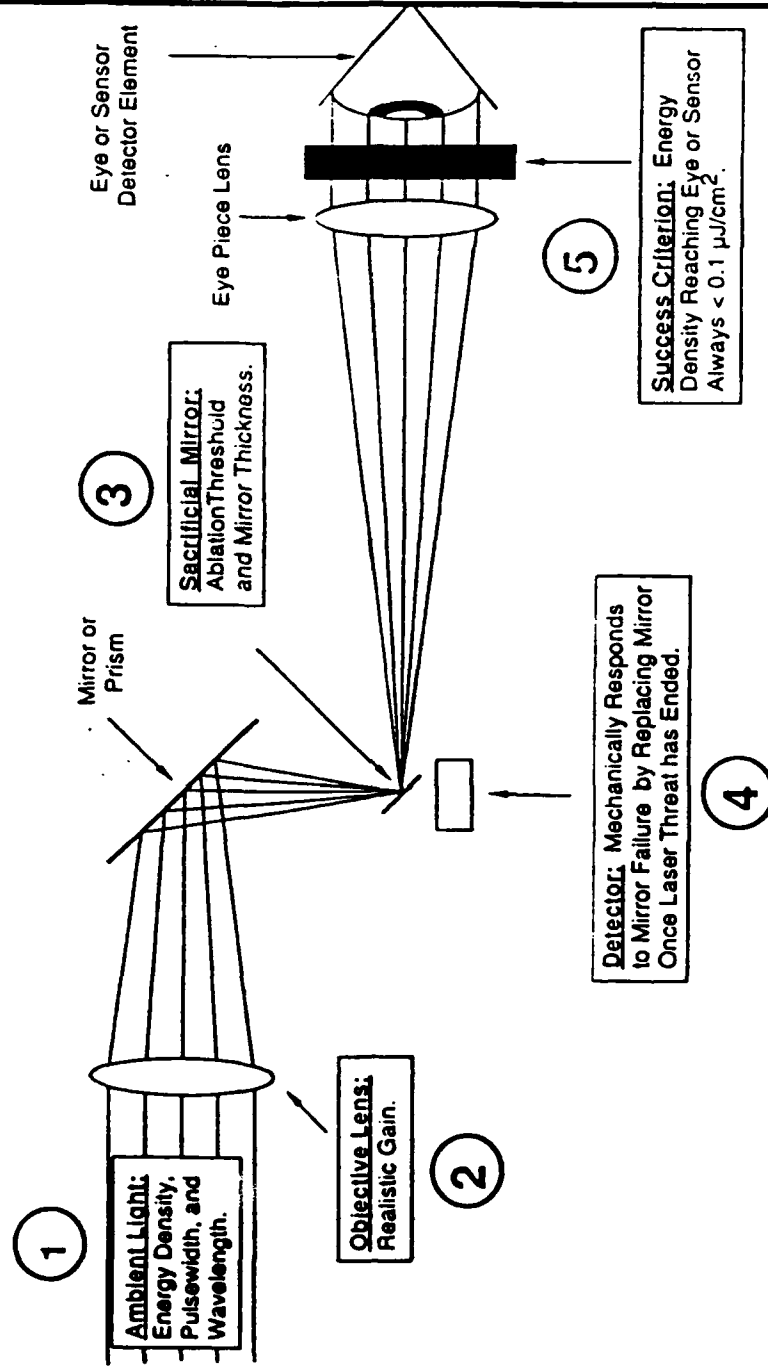


Figure 1b. A schematic diagram of the basic optical system for a mirror/optical-fuse device detailing important component operating parameters.

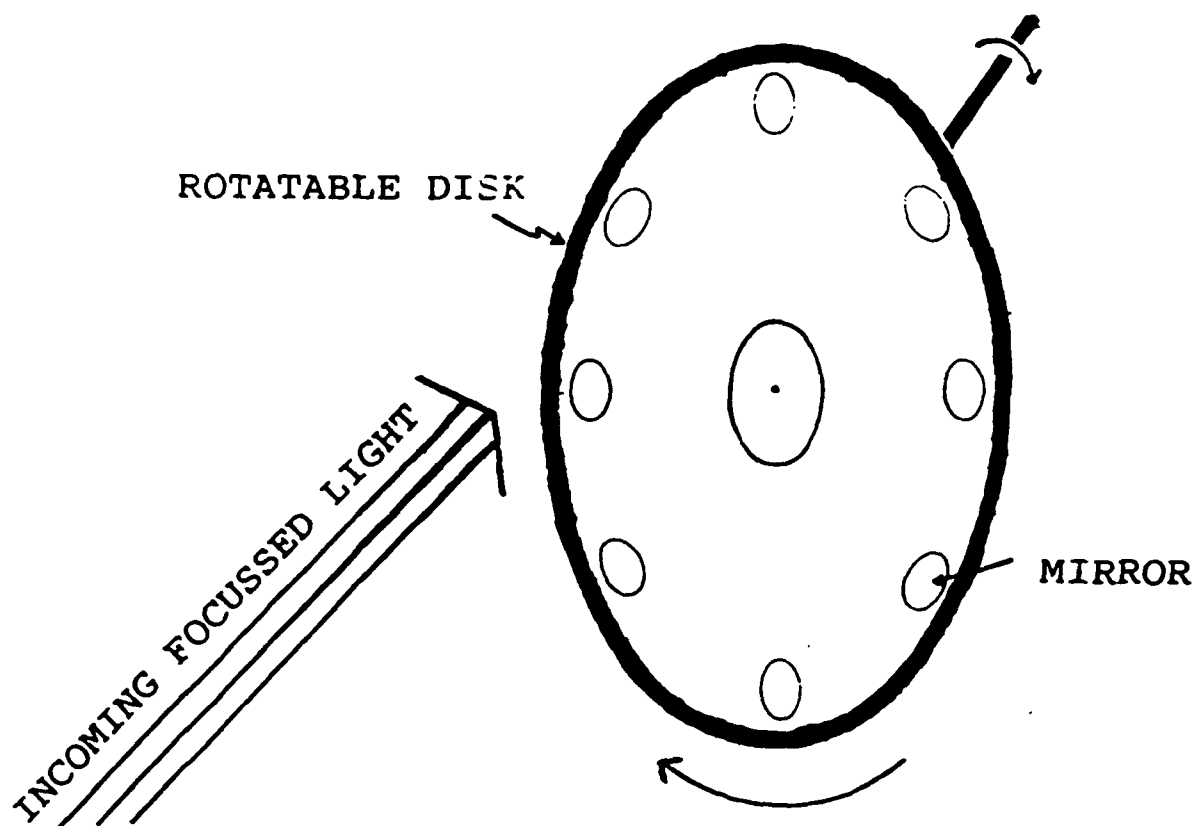


Figure 2. A sketch of one mirror/substrate design which would allow for easy and rapid repositioning of a new mirror surface after irradiation damage.

TABLE 1

Metal	LIDT (J/cm ²)	Pulsewidth, γ (ns)	Wavelength, λ (μm)
Silver	60	100	10.6
Ag	370	2000	10.6
	220	100	3.8
	200	100	2.7
	11	9	1.06
	37	500	0.492
Copper	12	1.4	10.6
Cu	17	28	10.6
	69	50	10.6
	95	90	10.6
	70	100	10.6
	56	100	10.6
	60	100	10.6
	480	2000	10.6
	230	100	3.8
	190	100	2.7
	90	9	1.06
	2-14	20	1.06
	11	500	0.492
Gold	43	100	10.6
Au	21	100	10.6
	37	100	10.6
	275	2000	10.6
	138	100	3.8
	123	100	2.7
	6	9	1.06
	13	500	0.492
Molybdenum	8	100	10.6
Mo	370	2000	10.6
	24	500	0.492
Aluminum	0.7	1.4	10.6
Al	40	100	10.6
	14	100	10.6
	8	500	0.492

Table 1. Measured laser-induced damage thresholds (LIDT) for optically pure common mirror materials.⁴⁻¹⁰ Thresholds may be lowered by controlled optical contamination with subsequent increase in absorption.